

NO DRAWINGS



Date of Application and filing Complete Specification: June 11, 1959.

No. 20086/59.

Application made in United States of America on June 30, 1958.

Complete Specification Published: Sept. 28, 1960.

Index at Acceptance:—Class 2(7), T(2X : 6B).

International Classification:—C08g.

COMPLETE SPECIFICATION

Organosiloxane gels

We, MIDLAND SILICONES LIMITED, a British Company of 68, Knightsbridge, London, S.W.1., do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to organosiloxane gels having unique non-flowing but self-healing properties, a process for the preparation thereof, and electrical apparatus potted or encapsulated therein.

A wide variety of materials have been used in the past for the so-called "potting", "filling" or "encapsulating" of electrical assemblies. Such materials have ranged from liquids to resinous or rubbery products in form, and are used to provide electrical insulation along with varying degrees of protection from thermal or mechanical abuse. Materials used in this manner in the past have generally not permitted visual inspection of the embedded apparatus, and except where they were actually liquids and hence subject to leakage from their container, they did not permit the electrical testing of components of the apparatus without rupture of the potting compound itself.

It is an object of this invention to provide a potting compound having optical clarity, a liquid nature prior to cure and a soft, tacky, non-friable, jelly-like consistency after being cured. A further object is to provide a material which, although non-flowing in the sense that it will not flow from a container, is self-healing to the withdrawal of a probe, and which has good dielectric properties as well as moisture and thermal resistance.

The above objects have been attained by the organosiloxane gel of the present invention, which can be defined as the reaction product

of an intimate mixture consisting essentially of (1) an organosiloxane having a viscosity of from 100 to 10,000 cs. at 25°C. and being a co-polymer consisting essentially of units of the general formulae $R\text{ViSiO}$, $R_2\text{SiO}$, and $\text{CH}_3\text{R}_2\text{SiO}_{0.5}$ where each R is a methyl or phenyl radical and Vi is a vinyl radical, at least 0.174 molar per cent of the units in said co-polymer being the said $R\text{ViSiO}$ units, (2) a liquid hydrogenosiloxane of the average general formula $\text{HRCH}_2\text{SiO}(\text{R}_2\text{SiO})_n\text{SiCH}_2\text{RH}$ where each R is as above defined and n has an average value such that the viscosity of the hydrogenosiloxane is not more than 10,000 cs. at 25°C., not more than 25 molar per cent of the total R radicals present in (1) and (2) being phenyl, and (3) a platinum catalyst in an amount sufficient to furnish at least 0.1 part per million of platinum calculated on the sum of the weights of (1) and (2); the proportions of (1) and (2) being such that prior to reaction there is an average of from 1.4 to 1.8 of the silicon-bonded H atoms in (2) per molecule of (1) and there being at least one $R\text{ViSiO}$ unit in (1) for every silicon-bonded H atom in (2), the molecular weight of (1) being calculated by the equation: $\log \text{visc.} = 1.00 + 0.0123 M^{0.5}$, where M is the molecular weight and "visc." is the viscosity in cs. at 25°C. of siloxane (1).

The reaction which takes place when the above defined materials stand in intimate contact with one another does not evolve gaseous by-products, and hence there are no voids in the gelled reaction product. The final product is a true gel, and it is insoluble in the common organic solvents. It has a rare combination of cohesive strength, adhesive properties, elasticity, and non fluidity which renders it completely non-flowing in respect to the container in which it has been gelled,

and which permits deformation under slight pressure. A probe can be inserted into or through the gel with great ease, so that electrical measurements can be taken upon any components which have been encapsulated therein. When such a probe is removed, the "self-healing" character of the gel is such that there is an immediate "flow" into the space formerly occupied by the probe, and there is no evidence of any rupture in the gel.

The combination of non-fluidity (in the one sense) with a self-healing type of flow, along with complete optical clarity and dielectric properties, makes the material eminently suitable for the filling, potting, encapsulating, or impregnating of electronic assemblies, capacitors, condensers, magnetic devices, or any other desired electrical apparatus.

The organosiloxane co-polymers defined as constituent (1) above are well-known materials. They can be prepared, for example, by the co-hydrolysis and co-condensation of the corresponding halosilanes, i.e. $R\text{ViSiCl}_2$, $R_2\text{SiCl}_2$, and $\text{CH}_3\text{R}_2\text{SiCl}$, or by the co-polymerisation and equilibration of the corresponding siloxanes. Thus, for example, siloxanes of the formulae $(R\text{ViSiO})_4$, $(R_2\text{SiO})_4$, and $(\text{CH}_3\text{R}_2\text{Si})_2\text{O}$ can be mixed in appropriate ratios and heated at 150° to 160°C . in the presence of a catalyst such as sodium hydroxide, potassium hydroxide and lithium hydroxide until an equilibrium is established, then the co-polymer neutralised with $\text{CH}_3\text{R}_2\text{SiCl}$. It is preferred that the co-polymer used in this invention be substantially free from silicon-bonded hydroxy groups.

The R radicals in the defined co-polymer can be the same or different radicals in each polymeric unit or in the molecule. Thus the co-polymer can contain the units MeViSiO and PhViSiO ; Me_2SiO , Ph_2SiO , and PhMeSiO ; and $\text{Me}_3\text{SiO}_{0.5}$, $\text{MePh}_2\text{SiO}_{0.5}$, and $\text{Me}_2\text{PhSiO}_{0.5}$ in any combination so long as a representative of each type is present and so long as the viscosity and vinyl content requirements are met and the phenyl content does not exceed 25 molar per cent. The symbols Me, Ph and Vi are used herein as meaning methyl, phenyl, and vinyl radicals respectively.

The co-polymer (1) should have a viscosity of from 100 to 10,000 cs. at 25°C . This is of course controlled by the amount of the end-blocking $\text{CH}_3\text{R}_2\text{SiO}_{0.5}$ units present. Preferably this co-polymer is substantially free from volatile low molecular weight species. As is well known, however, material of any particular viscosity will itself be composed of innumerable species of molecules having different molecular weights, and it is the viscosity of the mixture of species which is important here. Viscosities of from about 400 to 5000 cs. at 25°C . are preferred.

The hydrogenosiloxane (2) employed herein has the average general formula HRMeSiO (2) $(R_2\text{SiO})_n\text{SiMeRH}$, where each R is methyl or

phenyl and n is 0 or any positive integer or fraction so long as the viscosity does not exceed 10,000 cs. at 25°C . Thus n can vary from 0 to about 800 inclusive, the upper limit of course varying with the type of R radicals present. Viscosities in the range of from 2 to 2,000 cs. at 25°C . are most preferred. The R radicals in a given molecule or in a given mixture of molecular species falling within this definition can be the same or different radicals. The end-blocking units can be $\text{HMe}_2\text{SiO}_{0.5}$ units or $\text{HMePhSiO}_{0.5}$ units or both types. The repeating units which can be present can be Me_2SiO , MePhSiO , Ph_2SiO units, or a combination of such units. Any combination of these end-blocking and repeating units can be used. However, when phenyl radicals are present in either constituent (1) or constituent (2) as defined herein, the total number of such phenyl radicals should not exceed 25 molar per cent of the total R radicals present in (1) plus (2), with a preferred maximum being about 10 molar per cent. It is also preferred that the hydrogenosiloxane be substantially free from silicon-bonded hydroxy groups.

The defined hydrogenosiloxanes are known compounds and can be prepared, for example, by the co-hydrolysis and co-condensation of the corresponding chlorosilanes (i.e. MeRSiHCl and R_2SiCl_2) or by the acid catalysed equilibration of the siloxanes $(\text{MeRHSi})_2\text{O}$ and $(R_2\text{SiO})_4$.

The most important factor in the production of the gel of this invention is in the control of the proportions of the siloxane (1) and (2) employed. The proportions are such that prior to reaction there is an average of from 1.4 to 1.8 inclusive (preferably 1.45 to 1.7) of the silicon-bonded hydrogen atoms in (2) per molecule of (1), with there being at least one $R\text{ViSiO}$ unit in (1) for each such hydrogen atom, when the molecular weight (M) of siloxane (1) has been calculated by the equation: $\log \text{visc. (cs. at } 25^\circ\text{C.}) = 1.00 + 0.0123 M^{0.5}$. The unique properties of the gel of this invention are not obtained when proportions outside this definition are employed. The amount of "SiH" present in (2) is preferably determined by the known analytical methods for such determinations, although it too can be calculated from the viscosity of (2).

It can be seen that the weight ratios of (1) and (2) are thus subject to extremely wide variations, for they are dependent entirely upon the molecular weight of the one and the SiH content of the other. The equation used for determining the molecular weight of (1) gives a "number average" molecular weight, and has been shown by Dr. A. J. Barry to be reasonably valid for linear methylpolysiloxane liquids having molecular weights above 250 (Journal of Applied Physics, Vol. 17, 1020-1024, December, 1946). Of course, the expression may not be exactly accurate, particularly 130

when phenyl radicals or a relatively large amount of vinyl radicals are present. Nevertheless the calculation of molecular weight in this manner when used in conjunction with the defined limitations does express the proportions of (1) and (2) required for the desired results.

The required range of 1.4 to 1.8 SiH in siloxane (2) per molecular of (1) holds true regardless of the vinyl content of (1) so long as there is at least one RViSiO unit for each SiH. Thus no maximum limit of RViSiO content need be set, for an excess of vinyl radicals over silicon-bonded hydrogen atoms does not effect the peculiar non-flowing but self-healing characteristics of the gel. The thermal resistance of the gel, however, is somewhat adversely affected by large excesses of vinyl content. Thus it is preferred that there be not more than about 5 molar per cent of the RViSiO units present in the co-polymer (1). For optimum properties in general it is also preferred that substantially all the R radicals in both (1) and (2) be methyl radicals.

The gels of this invention are formed when the defined proportions of (1) and (2) are intimately mixed with a platinum catalyst and allowed to react. Many types of platinum catalysts for the SiH addition reaction to silicon-bonded vinyl radicals are known, ranging from platinum as such or as deposited on carriers such as silica gel or powdered charcoal, to platonic chloride, chloroplatinic acid, and salts of platinum. Any of such catalysts can be used in bringing about the reaction of the siloxanes (1) and (2) to form the unique gels of this invention. When optical clarity is an important factor, however, it is obvious that platinum in forms such as platinum black or platinised carbon or silk should be avoided. The preferred forms of catalyst are platonic chloride, platinum sulphate, salts of chloroplatinous acid such as K_2PtCl_4 and Na_2PtCl_4 , and chloroplatinic acid. The latter is the most potent form known, and hence is most preferred because at a given temperature a maximum reaction rate at a minimum concentration is obtained.

Chloroplatinic acid is most economically obtained as the hexahydrate $H_2PtCl_6 \cdot 6H_2O$, and this is the preferred form in this invention, although the dehydrated form is operative. It is important that the catalyst be intimately dispersed in the mixture, thus when the catalyst is one which is soluble in an inert solvent it is preferred to employ a solution of the catalyst as an aid to easy dispersion. Chloroplatinic acid is soluble in a number of organic solvents, e.g. glycols and esters, of which dimethyl phthalate and dimethyl carbitol are preferred examples.

The reaction between siloxanes (1) and (2) can take place at room temperature, or even below so long as the fluids remain in a liquid state. The reaction rate will of course vary

with the type and concentration of catalyst present, as well as with the temperature of the system. For example, with chloroplatinic acid as the catalyst in an amount to provide from 3 to 5 parts per million of platinum calculated on the sum of the weights of (1) and (2), the gel stage will be reached in from 2 to 6 days at about 25°C. Ordinarily it will be preferred to expedite the reaction by heating the mixture after it has been placed in position, and for a maximum reaction rate coupled with a minimum degree of expansion of the final gel it is preferred to heat the mixture at 125° to 165°C. At 3 to 5 ppm. of platinum furnished by chloroplatinic acid, the desired gel is obtained in less than one hour at 150°C. Since optimum physical properties are obtained by heat curing the gel after it has formed, it is most preferred to heat the system for about 8 hours at 125° to 150°C.

There should be at least 0.1 part per million of platinum present in the mixture, but for more practical rate of reaction it is preferred to use a minimum amount of 0.5 part per million. Since impurities in the system may stop the effective action of such trace amounts of catalyst, however, it is best to use in the range of 3 to 7 ppm. of platinum. An excess of catalyst does not affect the reaction adversely, so no particular maximum need be specified. However, for maximum optical clarity it is preferred to use a maximum of about 25 ppm. of platinum, even when a soluble catalyst such as chloroplatinic acid is the source. Considerations of economy will suggest the use of small amounts of catalyst regardless of whether a particular use of the gel requires optical clarity.

When chloroplatinic acid is the catalyst being used, it is best for purposes of shipping and storing the material to mix the desired amount of catalyst in the vinyl-containing co-polymer (1). Such a mixture is then diluted with the required amount of (2) just prior to using the final blend. It is not desirable to mix this particular catalyst with the SiH-containing siloxane (2) when the latter is to stand for a considerable time prior to use, for water may get into the system and cause a slow conversion of the SiH to SiOH groups.

Conventional techniques for potting, filling, or encapsulating electrical apparatus can be used with the material defined herein. After the defined mixture has been prepared, it can be positioned by pouring, dipping, spraying or any other suitable method. The mixture is then merely maintained in position, i.e. in contact with the apparatus in question, and either heated or allowed to stand until the mixture has reacted to form the desired gel.

The following examples illustrate the invention. All parts and percentages are by weight unless otherwise specified.

Example 1.

A series of mixtures was prepared, each

containing a particular co-polymer of Me_2SiO , MeViSiO , and $\text{Me}_3\text{SiO}_{0.5}$ units, and a particular hydrogenosiloxane of the general formula $\text{Me}_2\text{HSiO}(\text{Me}_2\text{SiO})_n\text{SiMe}_2\text{H}$. A solution of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$ in dimethyl phthalate was added to each mixture in an amount to provide about 4 parts per million of platinum calculated on the sum of the weights of the two siloxanes in each mixture. Samples of each mixture were then heated at 150°C . for 1 hour, and properties of the resulting gels were determined.

The types and amounts of siloxanes in each mixture are shown in Table I below. The co-polymer containing MeViSiO units is designated fluid (1), and that which contained the silicon-bonded H atoms is designated fluid (2). For purpose of comparison, mixtures both within and outside the scope of this invention are included in the table. Those gels which had the necessary degree of softness, tackiness, elasticity, and cohesiveness to be non-flowing and yet self-healing as defined above are labelled "good". Gels which were too soft or too hard to meet these requirements are labelled accordingly.

In the table, the viscosity of both fluids in cs . at 25°C . is listed under "visc." Under fluid (1), the molecular weight of that fluid was

calculated from the expression: $[(\log \text{visc.} - 1) + 0.0123]^2$. The "Pct. Vi" expresses the molar per cent of MeViSiO units present in fluid (1), and "Si/Mol (1)" shows the approximate average number of Si atoms (i.e. polymer units) per molecule of fluid (1). Since the amount of MeViSiO units present was relatively small, the Si/Mol. (1) value was approximated by merely dividing the molecular weight by 74, the unit weight of Me_2SiO .

Under fluid (2), "Wt. Pct." expresses the per cent of the fluid in the combined fluids, e.g. a value of 2.7 means 2.7 parts of fluid (2) per 97.3 parts of fluid (1). The per cent of silicon-bonded H atoms is designated "Pct. SiH" and was determined as a weight per cent of fluid (2) by direct analytical methods.

The controlling ratio in delineating the desired types of gels, as has been noted previously, is the ratio of silicon-bonded H atoms in (2) per molecular of (1). This ratio is shown under "SiH/Mol (1)" in the table. It is calculated directly from the per cent of fluid present, the per cent silicon-bonded H in that fluid, the molecular weight of (1), and the per cent of (1) present. Thus in Mix No. 1, the value would be calculated:

$$0.027 \times 0.00248 \times 22,800/0.973 = 1.57.$$

TABLE

Mix No.	FLUID (1)				FLUID (2)			RATIOS			Gel Quality
	Visc.	Mol. Wt.	Pct. Si/Mol Vi (1)		Wt. Pct.	Pct. SiH	Visc.	SiH/Mol (1)	SiH/Si (1)	SiH/Vi	
1	720	22,800	1.0	308	2.70	0.248	4.6	1.57	0.0051	0.51	Good
2	680	22,200	1.0	300	2.70	do.	do.	1.53	0.0051	0.51	Good
3	487	18,820	2.0	255	3.40	do.	do.	1.65	0.0066	0.33	Good
4	620	21,300	0.75	288	3.20	do.	do.	1.75	0.0061	0.81	Good
5	4490	46,300	2.0	627	1.37	do.	do.	1.59	0.0025	0.13	Good
6	4000	44,700	0.3	603	1.30	do.	do.	1.46	0.0024	0.8	Good
7	4000	44,700	1.0	603	1.30	do.	do.	1.46	0.0024	0.24	Good
8	720	22,800	1.0	308	36.03	0.0127	1018	1.63	0.0053	0.53	Good
9	680	22,200	1.0	300	4.76	0.139	40.6	1.53	0.0051	0.51	Good
10	720	22,800	1.0	308	2.26	0.248	4.6	1.31	0.0042	0.42	Soft
11	do.	do.	do.	do.	2.80	do.	do.	1.63	0.0053	0.53	Good
12	do.	do.	do.	do.	3.34	do.	do.	1.96	0.0063	0.63	Hard
13	487	18,820	2.0	255	2.80	do.	do.	1.35	0.0053	0.26	Soft
14	do.	do.	do.	do.	3.47	do.	do.	1.68	0.0066	0.33	Good
15	do.	do.	do.	do.	4.13	do.	do.	2.10	0.0079	0.39	Hard
16	4490	46,300	2.0	627	1.37	do.	do.	1.59	0.0025	0.13	Good
17	do.	do.	do.	do.	1.10	do.	do.	1.28	0.0020	0.10	Soft
18	do.	do.	do.	do.	1.47	do.	do.	1.72	0.0027	0.14	Good

It will be seen from the above table that factors such as the weight per cent of fluid (2) present, the viscosities of the fluids, or differences in the SiH or vinyl content do not in themselves control the nature of the reaction product. This is also true of the ratio between "SiH/Mol (1)" and "Si/Mol (1)" [shown as "SiH/Si (1)" in the table], and between "SiH/Si (1)" and "Pct. Vi" [shown as "SiH/Vi"

in the table].

Example 2.

Mixtures identical to Mix No. 1 in Example 1 were prepared, except that the chloroplatinic acid was added in quantities varying from 0.6 to 21.4 parts per million of platinum calculated on the sum of the weights of fluids (1) and (2). These mixtures all gelled satisfactorily in one hour at 150°C ., although that

containing 21.4 ppm. of platinum was not as optically clear as those containing smaller amounts.

Example 3.

5 A mixture identical to that of Mix No. 1 in Example 1, containing the same amount of platinum added as chloroplatinic acid, was allowed to stand at room temperature for six days. A satisfactory non-flowing, self-healing gel 10 was obtained.

Example 4.

A mixture was prepared of 97.3 parts of a co-polymer of Me_2SiO , MeViSiO , and $\text{Me}_3\text{SiO}_{0.5}$ units, having a viscosity of 700 cs. 15 at 25°C . and a MeViSiO content of 10 molar per cent, with 2.7 parts of the fluid (2) used in Mix No. 1 of Example 1, giving a ratio of about 1.55 silicon-bonded hydrogen per molecule of (2). A quantity of chloroplatinic acid 20 sufficient to provide 10 ppm. of platinum was thoroughly dispersed in the mixture, then it was heated at 150°C . for one hour. A satisfactory gel was obtained, although it was less thermally stable than that of Mix No. 1 25 in Example 1.

Example 5.

When Mix No. 1 of Example 1 was duplicated, but using 20 ppm. of platinum catalyst added as K_2PtCl_4 , as platonic chloride, or as 30 platinum deposited on charcoal, heating the mixtures at 50°C . produced non-flowing self-healing gels, although they were of poor optical quality.

Example 6.

35 When a mixture was prepared containing 99.5 parts of a co-polymer of Me_2SiO , MeViSiO , and $\text{Me}_3\text{SiO}_{0.5}$ units (having a viscosity of 4,000 cs. at 25°C . and a MeViSiO content of 0.5 molar per cent) and 0.5 part of 40 $(\text{PhMeHSi})_2\text{O}$, the "SiH/Mol (1)" ratio was about 1.74. Heating this mixture at 150°C . after dispersing 5 ppm. of platinum (added as chloroplatinic acid) therein provided a satisfactory non-flowing self-healing gel.

45 *Example 7.*

Co-polymers were prepared containing $\text{Me}_3\text{SiO}_{0.5}$, Me_2SiO , MeViSiO , and either PhMeSiO or Ph_2SiO units respectively. Each contained 1 molar per cent of the MeViSiO 50 units and 2 molar per cent of the particular phenylsubstituted unit, and each had a viscosity of about 700 cs. When 97.3 parts of either of these liquid co-polymers was mixed with 2.7 parts of the fluid containing 0.248% of 55 hydrogen from Example 1 and 5 ppm. of platinum added as chloroplatinic acid, heating the mixture for one hour at 150°C . gave a gel comparable with that obtained in Example 1, Mix No. 1.

60 **WHAT WE CLAIM IS:**

1. A soft, non-friable gel which is the reaction product of an intimate mixture consisting essentially of (1) an organosiloxane having a viscosity of from 100 to 10,000 cs. 65 at 25°C . and being a co-polymer consisting

essentially of units of the general formulae RViSiO , R_2SiO , and $\text{CH}_3\text{R}_2\text{SiO}_{0.5}$ where each R is a methyl or phenyl radical and Vi is a vinyl radical, at least 0.174 molar per cent of the units in said co-polymer being the said RViSiO units (2) a liquid hydrogenosiloxane of the average general formula $\text{HRCH}_2\text{SiO}(\text{R}_2\text{SiO})_n\text{SiCH}_2\text{RH}$ where each R is as above defined and n has an average value such that the viscosity of the hydrogenosiloxane 75 is not more than 10,000 cs. at 25°C ., not more than 25 molar per cent of the total R radicals present in (1) and (2) being phenyl, and (3) a platinum catalyst in an amount sufficient to furnish at least 0.1 part per million 80 of platinum calculated on the sum of the weights of (1) and (2); the proportions of (1) and (2) being such that prior to reaction there is an average of from 1.4 to 1.8 of the silicon-bonded hydrogen atoms in (2) per molecule of 85 (1) and there being at least one RViSiO unit in (1) for every silicon-bonded hydrogen atom in (2), the molecular weight of (1) being calculated by the equation: $\log \text{visc.} = 1.00 + 0.0123 \text{ M}^{0.5}$, where M is the molecular 90 weight and "visc." is the viscosity of siloxane (1) in cs. at 25°C .

2. A gel as claimed in claim 1 wherein the platinum catalyst is chloroplatinic acid, platonic chloride, platinum sulphate, or a metal salt 95 of chloroplatinous acid.

3. A soft, tacky, non-friable, non-flowing, self-healing gel which is the reaction product of an intimate mixture consisting essentially of 100 (1) an organosiloxane being substantially free from silicon-bonded hydroxy groups, having a viscosity of from 100 to 10,000 cs. at 25°C ., and being a co-polymer consisting essentially of units of the formulae MeViSiO , Me_2SiO and $\text{Me}_3\text{SiO}_{0.5}$ where Me and Vi are methyl 105 and vinyl radicals respectively, there being from 0.174 to 5 inclusive molar per cent of the MeViSiO units in said co-polymer, (2) a liquid hydrogenosiloxane substantially free from silicon-bonded hydroxy groups and having 110 the average general formula $\text{HMe}_2\text{SiO}(\text{Me}_2\text{SiO})_n\text{SiMe}_2\text{H}$ where Me is a methyl radical and n has a value such that the viscosity of the hydrogenosiloxane is from 2 to 2,000 cs. at 25°C ., and (3) chloroplatinic acid in an amount 115 sufficient to provide from 0.5 to 25 parts per million of platinum calculated on the sum of the weights of (1) and (2); the proportions of (1) and (2) being such that there is an average of from 1.45 to 1.7 of the silicon-bonded hydrogen atoms in (2) per molecule of (1) and there 120 being at least one MeViSiO unit in (1) for every silicon-bonded hydrogen atom (2), the molecular weight of (1) being calculated by the equation: $\log \text{visc.} = 1.00 + 0.0123 \text{ M}_{0.5}$, where M is the molecular weight and "visc" 125 is the viscosity of (1) in cs. at 25°C .

4. A method for capsulating electrical apparatus which comprises preparing the mixture defined in any one of the preceding 130

claims and maintaining the mixture in contact with said electrical apparatus until the mixture has reacted to form a soft, non-friable, non-flowing gel.

- 5 5. A process for the preparation of a soft, non-friable, non-flowing gel which comprises reacting, by contacting in a liquid phase, (1) an organosiloxane having a viscosity of from 100 to 10,000 cs. at 25°C. and being a co-poly-
 10 mer consisting essentially of units of the general formulae $RViSiO$, R_2SiO , and $CH_3R_2SiO_{0.5}$ where each R is a methyl or phenyl radical and Vi is a vinyl radical, at least 0.174 molar per cent of the units in said co-polymer
 15 being the said $RViSiO$ units, with (2) a liquid hydrogenosiloxane of the average general formula $HRCH_3SiO(R_2SiO)_nSiCH_3RH$ where each R is as above defined and n has an average value of from 0 to 800 inclusive, the viscosity
 20 of the hydrogenosiloxane being not more than 10,000 cs. at 25°C. and not more than 25 molar per cent of the total R radicals present in (1) and (2) being phenyl, in the presence of (3) chloroplatinic acid in an amount sufficient to provide at least 0.1 part per million of platinum calculated on the sum of the weights of

(1) and (2); the proportions of (1) and (2) being such that there is an average of from 1.4 to 1.8 of the silicon-bonded hydrogen atoms in (2) per molecule of (1) and there
 30 being at least one $RViSiO$ unit in (1) for every silicon-bonded hydrogen atom in (2), the molecular weight of (1) being calculated by the equation: $\log \text{visc.} = 1.00 + 0.0123 M^{0.5}$, where M is the molecular weight and "visc." is the viscosity of (1) in cs. at 25°C. 35

6. A process as claimed in claim 5 wherein the organosiloxane (1) is a co-polymer of $MeViSiO$, Me_2SiO and $Me_3SiO_{0.5}$ units, and wherein the hydrogenosiloxane has a
 40 viscosity of from 2 to 2,000 cs. at 25°C. and has the average general formula $HMe_2SiO-(Me_2SiO)_nSiMe_2H$, Me and Vi being methyl and vinyl radicals respectively.

7. A gel as claimed in claim 1 substantially as described with reference to any one of the Examples. 45

ELKINGTON & FIFE
 Consulting Chemists and Chartered Patent
 Agents, Bank Chambers, 329, High Holborn, 50
 London, W.C.1.
 Agents for the Applicants

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☐ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.